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Resonance laser ionization of atoms for nuclear physics

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Abstract

The applications of the laser resonance ionization method in nuclear research are reviewed. Investigation of radioactive isotopes using resonance ionization techniques provides a valuable combination of high selectivity, efficiency and spectral resolution. The majority of radioactive ion beams produced at on-line isotope separator facilities profit from the selectivity and universal applicability of laser ion sources. Numerous ultra-sensitive and high-resolution techniques of laser spectroscopy based on resonance ionization of atoms have been developed for the study of rare and radioactive isotopes. A summary of ionization schemes applied to radioactive isotopes is given in table form.

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(Some figures may appear in colour only in the online journal)

1. Introduction

Over the 50 years since the invention of lasers, a great variety of laser-based methods has been developed and applied to research in different fields of science. Among the first laser applications were spectroscopy and laser probing of matter. A high spectral brightness of laser radiation combined with wavelength tunability determines its outstanding capability to excite quantum transitions in atoms and molecules very selectively and efficiently. A majority of the laser spectroscopy methods are based on this resonance laser-matter interaction. Such an interaction is also a key element of the laser resonance ionization of atoms, which was invented in the early days of the laser epoch and now provides extraordinary capabilities for spectroscopy of short-lived isotopes and manipulation with the radioactive ion beams produced at accelerators.

The resonance ionization process is a multi-step photon absorption leading to a final state above the ionization threshold which can be driven by pulsed lasers very efficiently. This was demonstrated first for rubidium atoms by Ambartsumyan *et al* (1971). The results of this experiment and general consideration for two-step photoionization were presented at the 1971 IEEE/OSA Conference on Laser

Engineering and Applications (Letokhov and Ambartsumyan 1971, Ambartsumyan and Letokhov 1972).

In the following years, multi-step laser resonance ionization was developed and studied in numerous laboratories. The application of this method to the separation of uranium isotopes attracted most attention at that time. In this context, increasing ionization efficiency was of particular importance. Ionization schemes with excitation of autoionizing and Rydberg states were demonstrated (Janes *et al* 1976, Bekov *et al* 1978, 1979, Ambartsumyan *et al* 1975). The larger cross sections of the last step transitions provide more efficient ionization of atoms. Depending on the structure of atomic levels, ionization schemes with one or more resonance transitions can be applied as illustrated by figure 1. Theoretical grounds of the multi-step resonance excitation and ionization of atoms as well as multiple applications of laser photoionization spectroscopy can be found in the books by Letokhov (1987) and Hurst and Payne (1988).

The method of laser resonance ionization of atoms offers an outstanding combination of selectivity and sensitivity. Based on resonance ionization a broad variety of techniques has been developed for ultra-sensitive detection of single atoms and rare isotopes (Hurst *et al* 1979, Balykin *et al* 1980,

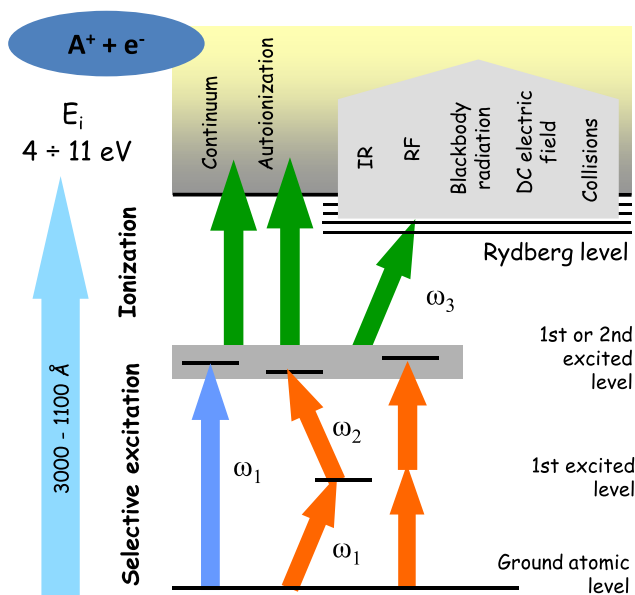


Figure 1. Schemes of resonance ionization.

Bekov and Letokhov 1983, Kudryavtsev 1992, Lu and Wendt 2003).

Contemporary nuclear physics explores the properties of short-lived isotopes which are available only in small amounts. The sensitive laser methods are playing an important role in establishing knowledge about nuclear matter, in particular for extracting properties such as nuclear spins, magnetic dipole moments, electric quadrupole moments and charge radii of nuclei (Kluge and Nörtershäuser 2003). The method of laser resonance ionization entered this field in 1983 (Alkhazov *et al* 1983) and since then it has kept a leading position in the sensitivity of optical spectroscopic research on radioactive isotopes.

At present, most of the short-lived radioactive isotopes are delivered in the form of ion beams by isotope separators on-line (ISOL) with an accelerator. In this approach isotopes are produced in a target under nuclear reactions initiated by high-energy projectile particles. Then products in ionic form are accelerated to some tens of keV and separated in magnetic fields according to their mass-to-charge ratio. Due to the presence of multiple isobars at a chosen mass, unambiguous isotope selection cannot be guaranteed by mass separation alone. The isobaric purity of the beam can be improved by the stage of ionization prior to its passage through the mass separator. Resonance ionization laser ion sources (RILIS) are implemented today at many ISOL facilities and demonstrate an outstanding performance due to their inherent selectivity combined with high ionization efficiency.

According to target and mass separator front-end conditions, two methods for the production of radioactive ion beams by resonance laser ionization have been successfully developed and operate nowadays at existing ISOL facilities:

- laser ionization of free atoms at low pressure in a hot cavity;
- laser ionization in a cell, filled with a buffer gas.

In section 2, we will overview these two types of ion sources and present the latest achievements in the development of these methods.

The applications of laser resonance ionization for the spectroscopy of radioactive isotopes will be discussed in section 3.

2. Laser ion sources for radioactive beams

2.1. Ionization in a hot cavity

The hot cavity concept has been developed with the goal of increasing the ionization efficiency of atoms moving in a vacuum through pulsed laser beams. The geometry of the cavity provides confinement of atoms within the laser beam during the time interval between consecutive laser pulses while the hot environment prevents atoms from absorption on the cavity internal walls. Several geometries have been proposed and they demonstrate an increase of ionization efficiency by high-pulse-repetition-rate lasers: a plane-parallel capacitor with an insulating cylinder and small holes (Andreev *et al* 1985, 1986a, b), a metallic cylinder with a small hole heated by an electron beam (Ames *et al* 1988, 1990), and a refractory metal tubular vessel with two small holes heated by dc current (Alkhazov 1989b, 1991), and a refractory metal tube (capillary) of internal diameter equal to the laser beam size (Fedoseev *et al* 1991, Mishin *et al* 1993). The last two configurations have been designed as ion sources for radioactive isotopic beams in conjunction with targets and mass separators at ISOL facilities IRIS (Investigation of Radioactive Isotopes on Synchrocyclotron) at the Petersburg Nuclear Physics Institute abbreviated as PNPI (Gatchina, Russia) and ISOLDE (Isotope Separator On-Line Detection Experiment) at the European Organization for Nuclear Research abbreviated as CERN (Geneva, Switzerland).

The nature of ion confinement and extraction in the hot cavity is identical to that of surface ion sources that have been used at ISOL facilities for a long time. Basic considerations for laser ionization in a tubular hot metal cavity were given by Mishin *et al* (1993) and by Fedoseyev *et al* (2000).

Diffusion of atoms out of a hot cavity ion source is relatively fast: depending on source dimensions and the thermal velocity of atoms, the time taken is about 0.1 ms. This delay is quite small in comparison to that of the target and ensures good conditions for ionization of short-lived isotopes. On the other hand, for efficient interaction of atoms with pulsed laser radiation, a pulse repetition rate of 10 kHz or more is required. Therefore the above-mentioned hot cavity ion sources were used with dye lasers pumped by copper vapour lasers (CVLs). CVLs are well suited to resonance ionization due to their short pulse duration (typically 15–30 ns), high average power (typically 30–40 W from one tube), convenient green and yellow radiation wavelengths and a flat power distribution across the beam providing good conditions for transverse pumping of dye lasers. The drawbacks of CVLs are long warm-up time, a complicated system of heat management, a high-voltage electrical circuit and the electromagnetic noise produced by high-voltage switching of the gas discharge in the laser tube.

However, a CVL system was operated successfully at ISOLDE RILIS from 1991 until 2008. In total, ion beams of 26 elements have been produced with this laser system at ISOLDE mass separators (Fedosseev *et al* 2008).

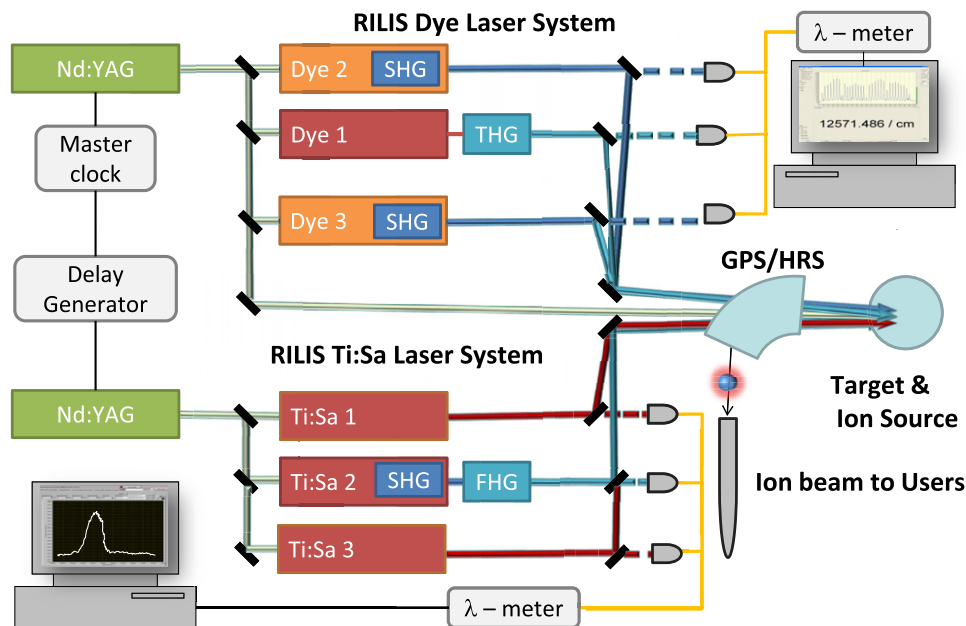


Figure 2. Layout of the RILIS setup at the ISOLDE facility at CERN. The second, third and fourth harmonics of laser beams can be produced with harmonics generation units SHG, THG and FHG, respectively.

At the IRIS facility a CVL system with dye lasers was put into operation in 1981. It was used for high-resolution resonance ionization spectroscopy of radioactive isotopes first in an atomic beam and then, since 1989, in a hot cavity ion source. Recently, a new CVL laser installation at the IRIS mass separator has been built (Barzakh *et al* 2012), and is capable of supplying laser beams of a much broader wavelength range. With that, the list of laser-ionized elements at IRIS has been extended beyond the rare-earths.

With the emergence of high-repetition-rate solid-state lasers, new promising alternatives to CVL and dye lasers have appeared. In particular, a system of titanium sapphire (Ti:Sa) lasers pumped by a Nd:YAG laser has been set up and operated with a pulse repetition rate of 6.6 kHz by Grüning *et al* (2004) at Mainz University (Germany) for resonance ionization mass spectrometry (RIMS) of plutonium. Ti:Sa-based laser systems have been further developed and chosen for the latest laser ion source projects at other ISOL facilities: Isotope Separation and ACceleration (ISAC) at TRI-University Meson Facility (TRIUMF), Vancouver, Canada (Lassen *et al* 2005), Holifield Radioactive Ion Beam Facility (HRIBF) at the Oak Ridge National Laboratory (ORNL), USA (Liu *et al* 2006), Ion Guide Isotope Separator On-Line (IGISOL) facility at University of Jyväskylä, Finland (Nieminen *et al* 2005), Système de Production d'Ions Radioactifs Accélérés en Ligne (SPIRAL1) and future SPIRAL2 facilities at Grand Accélérateur National d'Ions Lourds (GANIL) at Caen, France (Lecesne *et al* 2010). Extensive studies of ionization schemes for Ti:Sa lasers have been carried out in recent years by the LAsER Resonance Ionization Spectroscopy for Selective trace Analysis (LARISSA) group of Mainz University in collaboration with ORNL and TRIUMF. To date, 40 elements have been resonantly ionized using all solid-state laser systems.

At ISOLDE the hot cavity ion source RILIS is used for the majority of experiments. In order to substantially improve its operational conditions and to keep the possibility

of applying the already developed ionization schemes for the production of radioactive ion beams, it was decided to upgrade the laser system in several stages. In 2008–2009, the CVL system was replaced by a commercial Nd:YAG laser suitable for pumping the dye lasers. A q-switched solid-state laser based on the innovative slab (Innoslab) technology has been custom built for the RILIS installation by EdgeWave GmbH (Germany) in accordance with the required operating parameters. The laser generates pulses of ~ 10 ns duration at the rate of 10 kHz and delivers three output beams with user-controlled power distribution between them. Two beams at 532 nm (second harmonics of Nd:YAG laser) have a maximum combined average power of 100 W. Part of that power can be converted to the third harmonic (355 nm) with a maximum output of 20 W. The 355 nm beam enables pumping of laser dyes that emit light in the 372–550 nm range, which is an additional advantage of this new laser compared to CVL. Details of the laser configuration and comparison with parameters of formerly used CVLs are given by Marsh *et al* (2010).

In the next stages of the RILIS upgrade the old home-made dye lasers have been replaced by new commercial dye lasers and, finally, a complimentary fully solid-state Ti:Sa laser system has been installed.

Three Ti:Sa lasers and two newly designed frequency conversion units (Rothe *et al* 2011) have been constructed and installed at ISOLDE in collaboration with Mainz University. Figure 2 illustrates the new layout of the combined dye and Ti:Sa laser systems. By triggering both pump lasers from the same pulse and delay generator, it is possible to synchronize the pulses of the Ti:Sa and dye lasers so that successive steps of an atomic excitation scheme can be driven by a different type of laser. Beams of both laser systems can be transported to the ion sources of either of the two ISOLDE mass separators (General Purpose Separator (GPS) and High Resolution Separator (HRS)).

One of the main advantages of a metal hot cavity ion source is its robustness and resistance to mechanical and

therman stress, which is very important for operation in a highly radioactive environment typical of heavy targets of ISOL facilities. The high temperature conditions are indispensable for fast effusion of short-lived isotopes as well as for efficient ion storage and extraction. However, unwanted thermal ionization of atoms may take place on the hot surface, particularly if the ionization potential of the atom is low. The presence of surface-ionized isobars is the principal reason for selectivity reduction in hot cavity ion sources.

Different approaches to tackle this problem have been proposed and investigated. It was shown by Mishin *et al* (1993) and further investigated by Schwellnus *et al* (2009) that making the hot cavity of low-work-function materials does improve the selectivity of RILIS. Following that study, RILIS cavities made of GdB₆ (supplied by Huizhou Tian Yi Rare Material Co. Ltd, China) are being used for some experiments at ISOLDE which require more efficient suppression of isobars, in particular for isotopes of rare-earth elements.

With the goal of substantially improving the selectivity of RILIS, Blaum *et al* (2003) suggested performing laser ionization inside a linear radio-frequency quadrupole trap. This laser ion source and trap (LIST) in combination with a positively biased electrode could be installed closely to the outlet of the hot transfer tube. The electrode repels ions emerging from the tube, while only neutral species enter the trap and interact with laser beams. The LIST concept was developed at Mainz University and has been adapted to the conditions of the ISOLDE front end. Recently, a first on-line test of the LIST with a titanium-foil target was carried out at ISOLDE. Stable and radioactive magnesium isotopes were laser ionized, while surface-ionized aluminum, titanium and potassium isotopes were suppressed. The ion current of titanium isotopes versus the repelling voltage is shown in figure 3. The suppression factor for ions produced inside the hot cavity (the ratio of the ion current at negative repelling voltage to that at +10 V) deduced from this plot is 3000. Since the confinement of atomic flow within the laser beams does not work in the LIST, only a small fraction of magnesium atomic flow could interact with laser beams. The ionization efficiency for magnesium was therefore lower by a factor of 50. A good feature of this approach is that by changing the polarity of the repelling electrode, one can switch the system to a normal hot cavity RILIS with efficient laser ionization, ion extraction and guiding through the trap to the mass separator accelerating field region. Such a beam manipulation might be useful in certain cases.

Another possibility of reducing the fraction of surface-ionized ions in a beam produced by a hot cavity RILIS is based on the pulsed nature of laser ionization. If laser ions are extracted from the cavity quickly, they will form a bunch which can be time-gated out of the continuously produced surface ions. The ion beam gating synchronized to laser pulses was applied in some experiments with RILIS at ISOLDE (Jading *et al* 1997, Lettry *et al* 1998), but the improvement of selectivity did not exceed a factor of four because the extraction of ions from the cavity was relatively slow. A much improved selectivity could be achieved by applying the in-source time-of-flight technique proposed by Mishin *et al* (2009) for ion pulse compression. The technique has been demonstrated experimentally: combining a high

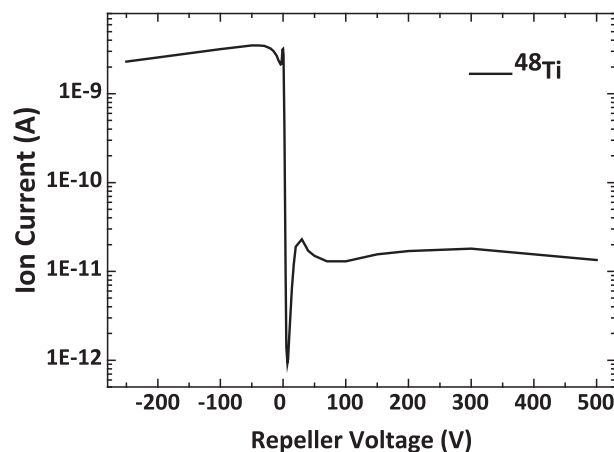


Figure 3. The ion current of titanium transmitted through the LIST versus the voltage applied to the repelling electrode located between the surface ion source and LIST.

electrical resistance of the ionizer graphite tube, pulsed heating current and an appropriate field-free drift space, ion bunches of laser-ionized thulium were compressed to a width of 5 μ s and separated in time from thermal ions. A selectivity improvement by a factor of 100 could be potentially attained employing this technique for a hot cavity RILIS without losing the ionization efficiency.

2.2. Ionization in a gas cell

The operational principle of the laser ion source is based on an element-selective resonance multi-step laser ionization of neutral atoms that, after production in a nuclear reaction, are thermalized and neutralized in a buffer gas where weak plasma is created by the primary accelerator beam, the recoil ions and the radioactivity. This method was developed at K U Leuven in the early 1990s (Kudryavtsev *et al* 1996, Van Duppen *et al* 1992, Vermeeren *et al* 1994), and has been used since then at the Leuven Isotope Separator On-Line (LISOL) facility at Louvain-La-Neuve (Belgium) to produce short-lived radioactive isotopes in different types of nuclear reactions. Recently, it was implemented at the IGISOL facility (Moore *et al* 2010) and in RIKEN (Rikagaku Kenkyusho, Institute of Physical and Chemical Research, Japan) (Sonoda *et al* 2011). Laser ionization spectroscopy in a gas cell in ‘off-line’ conditions has been developed as well at Mainz University (Backe *et al* 1997, Sewtz *et al* 2003). The extraction time of radioactive isotopes out of the gas-cell-based ion source is defined by the gas flow and is much shorter (down to 10 ms) in comparison to the hot cavity ion source and does not depend on chemical properties of extracted atoms that are especially important for refractory atoms.

The latest version of the gas-cell-based laser ion source for fusion–evaporation reactions with spatially separated stopping and laser ionization chambers is shown in figure 4 (Kudryavtsev *et al* 2009). The accelerator beam enters the cell through a thin molybdenum foil. The nuclear reaction products recoiling out of the target are thermalized and neutralized in a high-purity (<1 ppb) noble gas (helium or argon) at pressures of 100–500 mbar. The stopped recoils are brought from the stopping volume to the laser ionization

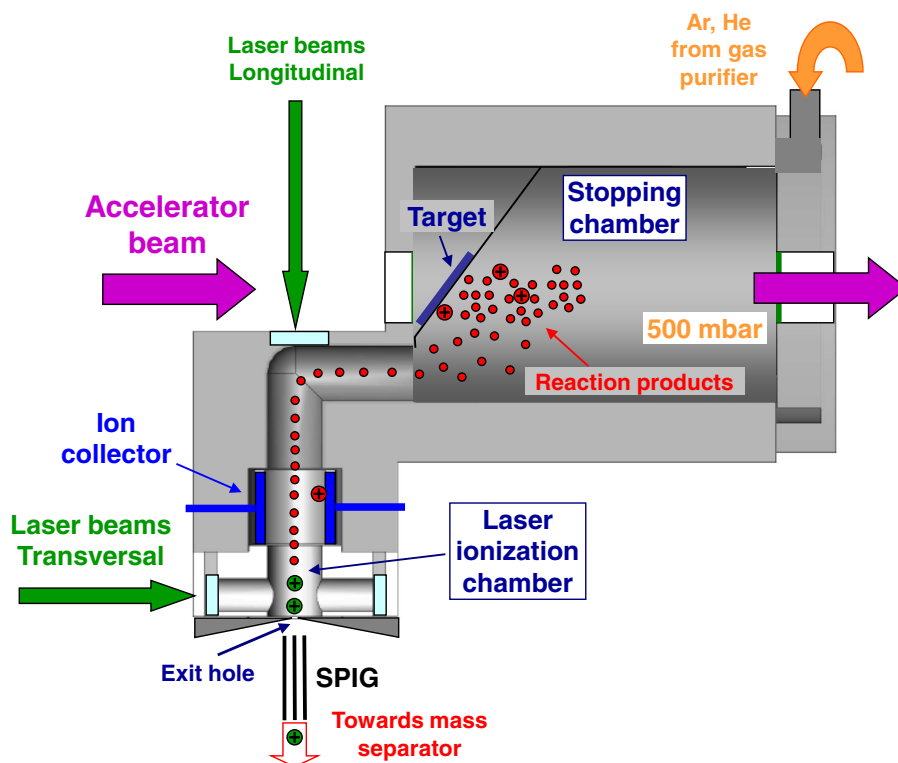


Figure 4. A schematic drawing of the dual-chamber gas-cell-based laser ion source.

volume by the gas flow. Non-neutralized ions are collected by an ion collector. A two-step two-colour scheme is used for the ionization of atoms of interest via autoionizing state. The pulsed laser beams with a repetition rate of 200 Hz enter the ionization chamber transversely and ionize atoms in the exit hole region. The evacuation time of the laser-irradiated volume is about 15 ms, with an exit hole diameter of 0.5 mm, which is longer than the time between two subsequent laser pulses of 5 ms guaranteeing that all atoms have been irradiated by laser light. Ions leaving the gas cell are captured by a sextupole ion guide (SPIG) (Van den Bergh *et al* 1997) and transported towards the mass separator.

The laser ionization of stable atoms created by evaporation from a filament inside the cell can be performed using a longitudinal beam path. This allows us to obtain information about the properties of the ion collector, evacuation of the gas cell, gas purity and formation of molecular ions from laser-produced ions in reactions with the noble gas and residual molecules. This is important for the ion source efficiency since the laser-produced ions can be lost in molecular side bands (Facina *et al* 2004, Kudryavtsev *et al* 2001). A very efficient way of avoiding the problem is to apply a voltage between the gas cell and the SPIG rods. In this case, weakly bound side-band molecular ions can be converted into atomic ions.

The laser ion source has been used for the production of exotic nuclei for nuclear spectroscopy studies in proton-induced fission and light- and heavy-ion-induced fusion evaporation reactions; see table 1. An example of the application of the cell for laser production of neutron-deficient isotopes in the region close to the $N = Z$ line is shown in figure 5 where gamma spectra on mass 94 detected in coincidence with emission of β -particles (β -gated) are

displayed. The radioactive ^{94}Rh isotopes were produced by impinging a ^{40}Ar beam on a ^{58}Ni target. The laser ion source selectivity, defined as the ratio of the ion count rate with lasers ON resonance to the one with lasers OFF, of more than 2200 has been obtained; see the caption of figure 5.

3. Resonance ionization laser spectroscopy of radioactive isotopes

3.1. Laser spectroscopy of mass-separated isotopes

The laser resonance ionization spectroscopy (RIS) is an extremely sensitive method of probing the nuclear ground and metastable states via measurements of isotopic shifts (IS) and hyperfine structure (HFS) of atomic transitions. Most commonly, a high level of spectral resolution is provided by using collimated beams of radioisotopes interacting with laser beams in orthogonal or collinear geometry.

In the first study of radioisotopes using the RIS method (Alkhazov *et al* 1983, Zherikhin *et al* 1984) samples of $^{145-150}\text{Eu}$ were produced at the IRIS ISOL facility, accumulated on tantalum foils in quantities 10^{10} – 10^{11} atoms and placed in the high-temperature source of the atomic beam. Laser beams generated by CVL-pumped dye lasers interacted with the atomic beam at a right angle. The IS measurements were carried out at the first step of a three-step ionization scheme by detecting photo-ions while scanning the laser frequency across the resonance. With the laser linewidth of 600 MHz the accuracy of IS measurements was equal to 80–100 MHz.

This ‘off-line’ operation was used only for isotopes with a relatively long lifetime (more than four days). For RIS of short-lived isotopes an ‘on-line’ technique was developed

Table 1. Radionuclides and radioactive ion beams delivered for atomic and nuclear physics experiments applying methods of resonance laser ionization. The information on the ionization scheme used includes a number of excitation steps and techniques (HC RILIS, hot cavity RILIS; GC RILIS, gas cell RILIS; ABT, atomic beam with thermal evaporation; ABPL, atomic beam with pulsed laser evaporation; CRIS, collinear resonance ionization spectroscopy; RIMS, resonance ionization mass spectroscopy), and the type of ionizing transition: C, continuum; A, autoionization; R, Rydberg state.

	Z	Scheme	A	Technique	Facility	Reference
Li	3	Four-step-C	8, 9	ABT	UNILAC/GSI	Ewald <i>et al</i> 2004
			8–11		ISAC/TRIUMF	Sánchez <i>et al</i> 2006
Be	4	Two-step-A	7, 10–12, 14	HC RILIS	ISOLDE/CERN	Köster <i>et al</i> 1998
		Three-step-C	10–12		ISAC/TRIUMF	Prime <i>et al</i> 2006
		Two-step-A	9–12			Lassen 2011
Mg	12	Three-step-C	23, 27–34	HC RILIS	ISOLDE/CERN	Köster <i>et al</i> 2003b
			22			Mukherjee <i>et al</i> 2004
			21			Krämer <i>et al</i> 2009
		Three-step-C	21, 23, 27, 28		ISAC/TRIUMF	Lassen <i>et al</i> 2009
		Three-step-A	20, 21, 23–35			Lassen 2011
Al	13	Two-step-C	26, 28–34	HC RILIS	ISOLDE/CERN	Köster <i>et al</i> 2003b
		Two-step-C	26		ISAC/TRIUMF	Prime <i>et al</i> 2006
			26, 28, 29			Lassen <i>et al</i> 2009
			30–31			Lassen 2011
Ca	20	Three-step-C	49–52	HC RILIS	ISAC/TRIUMF	Lassen 2011
Mn	25	Three-step-A	53, 54, 56–65	HC RILIS	ISOLDE/CERN	Fedoseyev <i>et al</i> 1997
			61–69			Hannawald <i>et al</i> 1999
			48–54, 56–69			Oinonen <i>et al</i> 2000
Fe	26	Two-step-A	65, 67	GC RILIS	LISOL/LLN	Pauwels <i>et al</i> 2009
Co	27	Two-step-A	66, 68, 70	GC RILIS	LISOL/LLN	Mueller <i>et al</i> 2000
			67			Weissman <i>et al</i> 1999
			65, 67			Pauwels <i>et al</i> 2009
Ni	28	Three-step-A	56, 57, 59, 63, 65–70	HC RILIS	ISOLDE/CERN	Jokinen <i>et al</i> 1997
		Two-step-A	54, 55	GC RILIS	LISOL/LLN	Reusen <i>et al</i> 1999
		Two-step-A	68–74			Franchoo <i>et al</i> 2001
Cu	29	Two-step-A	57–62, 64, 66–78	HC RILIS	ISOLDE/CERN	Köster <i>et al</i> 2000b
		Two-step-A	57–59	GC RILIS	LISOL/LLN	Cocolios <i>et al</i> 2010
		Two-step-A	70–76			Kruglov <i>et al</i> 2002
Zn	30	Three-step-C	58	HC RILIS	ISOLDE/CERN	Jokinen <i>et al</i> 1998
			58–61, 63			Oinonen <i>et al</i> 2000
			58–63, 69, 71–74			Köster <i>et al</i> 2003
			61–63, 65, 69, 71–81			Köster <i>et al</i> 2005
			71–82			Köster <i>et al</i> 2008
Ga	31	Two-step-C	61	HC RILIS	ISOLDE/CERN	Weissman <i>et al</i> 2002b
			74–86			Köster 2002a
			61–68, 70, 72–75			Köster <i>et al</i> 2003a
		Two-step-R	62		ISAC/TRIUMF	Prime <i>et al</i> 2006
		Two-step-C	61–68, 70, 72–75			Lassen <i>et al</i> 2009
Ge	32	Three-step-A	67, 71, 76	HC RILIS	ISAC/TRIUMF	Lassen 2011
Sr	38	One-step-R	89, 90	CRIS	Mainz University	Monz <i>et al</i> 1993
Tc	43	Three-step-A	94, 96, 98, 99	HC RILIS	ISAC/TRIUMF	Lassen 2011
Ru	44	Two-step-A	90, 91	GC RILIS	LISOL/LLN	Dean <i>et al</i> 2004
Rh	45	Two-step-A	91–93	GC RILIS	LISOL/LLN	Dean <i>et al</i> 2004
		Two-step-A	116			Kudryavtsev <i>et al</i> 2009
Ag	47	Three-step-C	112, 121–127	HC RILIS	ISOLDE/CERN	Fedoseyev <i>et al</i> 1995b
			107m, 122–129			Kratz <i>et al</i> 1998
			101–108, 110–129	HC RILIS	ISAC/TRIUMF	Fedoseyev <i>et al</i> 2000
		Three-step-C	129–130			Kratz <i>et al</i> 2005
			98–107, 109–117			Lassen <i>et al</i> 2009
		Two-step-C	97–101	GC RILIS	LISOL/LLN	Darby (to be published)
Cd	48	Three-step-C	131, 132	HC RILIS	ISOLDE/CERN	Hannawald <i>et al</i> 2000
			98–105, 107, 109, 111, 115, 117–132			Köster <i>et al</i> 2003b
			129–133			Kratz <i>et al</i> 2005
In	49	Two-step-C	100–108	HC RILIS	ISOLDE/CERN	Köster 2002a
			132–135			Dillmann <i>et al</i> 2002
		Two-step-C	108, 110, 112, 116		ISAC/TRIUMF	Lassen 2011
Sn	50	Three-step-A	101–103, 108	HC RILIS	UNILAC/GSI	Fedoseyev <i>et al</i> 1995a
			109–111, 113, 117, 119, 121, 123, 125–137		ISOLDE/CERN	Fedoseyev <i>et al</i> 2000
			136–138			Walters <i>et al</i> 2005
			105–110, 113, 117, 119, 121, 123, 125, 128–138			Köster <i>et al</i> 2008
		Two-step-A	125–132	ABPL		Le Blanc <i>et al</i> 2002
		Three-step-A	107–111, 113, 121	HC RILIS	ISAC/TRIUMF	Lassen 2011

Table 1. (Continued.)

	Z	Scheme	A	Technique	Facility	Reference
Sb	51	Three-step-C	128–138 137–139	HC RILIS	ISOLDE/CERN	Fedosseev <i>et al</i> 2008 Arndt <i>et al</i> 2012
Te	52	Three-step-C	120, 122–136	ABPL	ISOLDE/CERN	Sifi <i>et al</i> 2006
Pr	59	Three-step-C	136, 140	HC RILIS	ISOLDE/CERN	Gottberg 2011
Nd	60	Three-step-C	132, 134–141 138, 139, 140	ABT HC RILIS	IRIS/PNPI ISOLDE/CERN	Letokhov <i>et al</i> 1992 Gottberg 2011
Sm	62	Three-step-A	138–143, 145 140–143	ABT HC RILIS	IRIS/PNPI ISOLDE/CERN	Mishin <i>et al</i> 1987b Gottberg 2011
Eu	63	Three-step-A	145–150 141–144 155–159 138–145	ABT	IRIS/PNPI	Alkhazov <i>et al</i> 1983 Fedoseyev <i>et al</i> 1984 Alkhazov <i>et al</i> 1990a Letokhov <i>et al</i> 1992
Gd	64	Three-step-A	137–139, 141–144 146, 148, 150 143, 145, 146	HC RILIS ABT HC RILIS	IRIS/PNPI	Barzakh <i>et al</i> 2004 Alkhazov <i>et al</i> 1988 Barzakh <i>et al</i> 2005
Tb	65	Three-step-A	147–155, 157, 159 149	ABT HC RILIS	IRIS/PNPI ISOLDE/CERN	Alkhazov <i>et al</i> 1990b Köster <i>et al</i> 2003b
Dy	66	Three-step-C	151–155, 157, 159	HC RILIS	ISOLDE/CERN	Gottberg 2011
Ho	67	Three-step-A	152–163 152	ABT HC RILIS	IRIS/PNPI	Alkhazov <i>et al</i> 1989a Alkhazov <i>et al</i> 1991
Tm	69	Three-step-A	157–168, 170–172 153, 154, 158–163	ABT HC RILIS	IRIS/PNPI	Mishin <i>et al</i> 1987a Barzakh <i>et al</i> 2000
Yb	70	Two-step-R Three-step-A	157, 159, 175 154–156, 160–166 155 153–156, 158–164 157–167 178	CRIS HC RILIS	ISOLDE/CERN IRIS/PNPI	Schulz <i>et al</i> 1991 Alkhazov <i>et al</i> 1992 Barzakh <i>et al</i> 2000 Barzakh <i>et al</i> 2002 Mishin <i>et al</i> 1993 Gottberg 2011
Ir	77	Three-step-C	182–189	ABPL	ISOLDE/CERN	Verney <i>et al</i> 2000
Pt	78	Three-step-C	186, 188 185–189, 191 183–189, 191, 193	ABPL	ISOCELE/IPN	Lee <i>et al</i> 1988 Duong <i>et al</i> 1989 Hilberath <i>et al</i> 1992 Le Blanc <i>et al</i> 1999
Au	79	Three-step-A Three-step-C Three-step-A Two-step-C Three-step-C Three-step-A Three-step-C Two-step-C Three-step-A	178–185 185–189 195 194–196, 198 186, 187, 190, 192, 194–196 198, 199 183, 184 183–186, 191 184, 188, 190, 191, 193, 195 194 184, 186, 190, 194, 201, 202	ABT ABPL HC RILIS	ISOLDE/CERN McGill University ISOCELE/IPN ISOLDE/CERN ISOCELE/IPN ISOCELE/CERN	Wallmeroth <i>et al</i> 1987 Krönert <i>et al</i> 1987 Lee <i>et al</i> 1987a Lee <i>et al</i> 1987b Savard <i>et al</i> 1990 Krönert <i>et al</i> 1991 Le Blanc <i>et al</i> 1992 Sauvage <i>et al</i> 2000 Eliseev <i>et al</i> 2010 Podolyak 2010
Tl	81	Two-step-R Two-step-C	208 179–200 183–191, 193–195, 197, 207	GC RILIS HC RILIS	Mainz University ISOLDE/CERN	Lauth <i>et al</i> 1992 Köster <i>et al</i> 2003b Barzakh <i>et al</i> 2012
Pb	82	Three-step-C	185 184–203, 205, 209–215 183–203, 215 182–190	HC RILIS	ISOLDE/CERN	Andreyev <i>et al</i> 2002 Köster 2002b Köster <i>et al</i> 2003b Seliverstov <i>et al</i> 2006
Bi	83	Three-step-C	188–208, 210–218	HC RILIS	ISOLDE/CERN	Köster <i>et al</i> 2003b
Po	84	Three-step-C	193–198, 200, 202, 204 192–210, 216, 218 (even) 191–203, 209, 211 (odd)	HC RILIS	ISOLDE/CERN	Cocolios <i>et al</i> 2008 Cocolios <i>et al</i> 2011 Seliverstov <i>et al</i> 2012
At	85	Three-step-C Two-step-C Three-step-C Three-step-R	198, 199, 212, 217, 218 197–202 193–205, 217 205	HC RILIS	ISAC/TRIUMF ISOLDE/CERN	Lassen 2011 Rothe <i>et al</i> 2012
Fr	87	Two-step-C Two-step-R	221 221	HC RILIS	ISAS ^a , Troitsk	Andreev <i>et al</i> 1986b Andreev <i>et al</i> 1987
Ac	89	Two-step-A	212, 213 225	GC RILIS HC RILIS	LISOL/LLN ISAC/TRIUMF	to be published Lassen 2011
Th	90	Two-step-C Three-step-A	230 228–230	RIMS	LANL ^b , USA Mainz University	Johnson and Fearey 1993 Raeder <i>et al</i> 2011b
Np	93	Two-step-A Three-step-A	237	RIMS	Mainz University	Riegel <i>et al</i> 1993
Pu	94	Three-step-A	239–242, 244	RIMS	Mainz University	Ruster <i>et al</i> 1989
Am	95	Three-step-R	243	RIMS	Mainz University	Erdmann <i>et al</i> 1998

Table 1. (Continued.)

	Z	Scheme	A	Technique	Facility	Reference
		Two-step-C	241, 243, 240f, 242f 244f	GC RILIS	MPIK ^c , Heidelberg	Backe <i>et al</i> 1998 Backe <i>et al</i> 2000
Cm	96	Three-step-R	248	RIMS	Mainz University	Erdmann <i>et al</i> 1998
Bk	97	Three-step-R	249	RIMS	Mainz University	Erdmann <i>et al</i> 1998
Cf	98	Three-step-R	249	RIMS	Mainz University	Erdmann <i>et al</i> 1998
Es	99	Three-step-R	254	RIMS	Mainz University	Erdmann <i>et al</i> 1998
Fm	100	Two-step-C	255	GC RILIS	Mainz University	Sewtz <i>et al</i> 2003

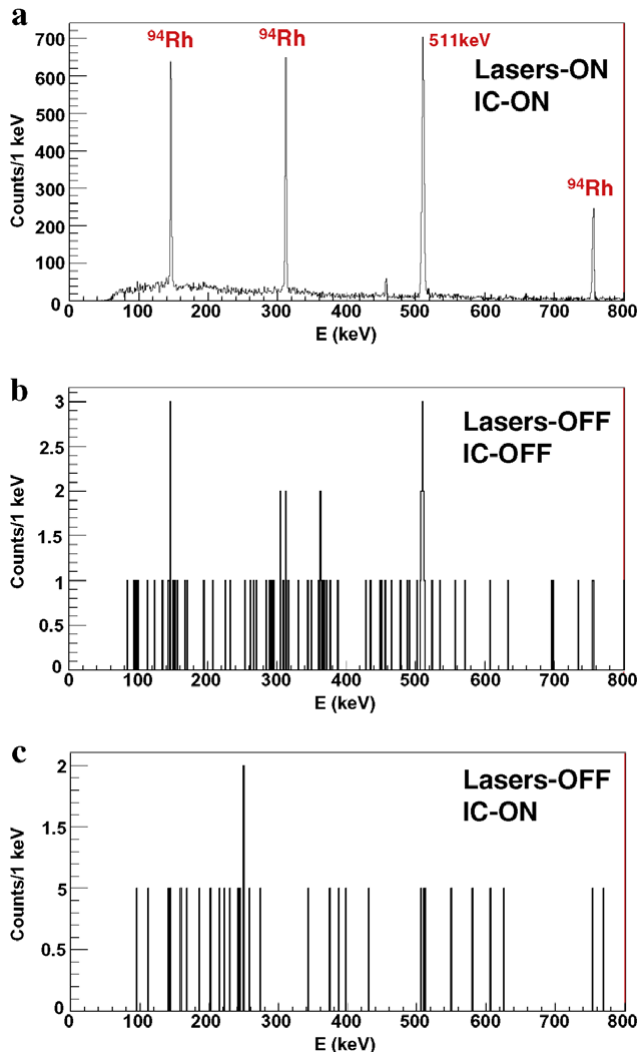
^aInstitute of Spectroscopy, Academy of Sciences of the USSR.^bLos Alamos National Laboratory.^cMax-Planck-Institut für Kernphysik.

Figure 5. β -gated gamma spectrum obtained at mass 94: (a) with lasers tuned in resonance to rhodium isotopes and IC—ON, (b) lasers—OFF and IC—OFF, (c) lasers—OFF and IC—ON. The measuring time is 300 s. The ^{94}Rh atoms were produced in the $^{40}\text{Ar} + ^{58}\text{Ni}$ heavy ion fusion evaporation reaction.

by Fedoseyev *et al* (1984). With that, a photoionization chamber was set at the output beam line of the IRIS mass separator in such a way that the ion beam was directed into the bottom of an atomic beam source (crucible) from which they could be immediately evaporated, collimated and ionized by laser beams. This development was accompanied by the implementation of a multi-pass mirror system for laser beams. The ionization efficiency has reached 3×10^{-4} , and

IS for isotopes $^{141-144}\text{Eu}$ with half-life periods in the range of 2.6 min–10 s have been measured. With this arrangement it was possible to boost up the flux of isotopes in the atomic beam by making accumulation of isotopes in a cold crucible. The temperature of the crucible was then quickly raised and a few laser scans with a higher signal/noise ratio were performed. Moreover, in many cases, this method was used to increase the quantity of the isotopes under investigation following the decay of parent nuclei in the cold crucible.

A substantial improvement of spectral resolution was achieved by applying a single-mode continuous wave (cw) dye laser for RIS of rare-earth isotopes at the IRIS facility (Mishin *et al* 1987a, b). The cw laser radiation was pulse-amplified in a dye cell pumped by a CVL beam. A spectral bandwidth of the amplified radiation equal to 50 MHz enabled measurements with a resolution of 150 MHz determined by the collimation geometry of the atomic beam.

This technique has been used in a number of experiments at the IRIS facility for the measurements of IS and HFS in isotopic chains of europium, thulium, samarium, neodymium, gadolinium, terbium and holmium (see table 1).

At the ISOLDE facility the laser resonance ionization was applied for the first time by Wallmeroth *et al* (1987) for the study of IS and HFS of $^{185-189}\text{Au}$ isotopes in an atomic beam geometry. Similarly to Fedoseyev *et al* (1984), the ion beam of radioisotopes was directed into a crucible, although the gold isotopes were obtained as daughters of abundantly produced and mass separated mercury isotopes after their decay in the crucible. Ions were detected with a time-of-flight mass spectrometer. The lasers were operated at a 10 Hz pulse rate; therefore the detection efficiency was quite low, 10^{-8} . A way to improve the efficiency of RIS in atomic beams using low-pulse-rate lasers was developed by Krönert *et al* (1987): they applied a pulsed-laser-induced desorption to evaporate accumulated radioisotopes to generate a pulsed atomic beam with time structure adequate for efficient resonance ionization. In parallel with ISOLDE, the same approach was developed at the McGill University (Montréal, Canada) and Institut de Physique Nucléaire d'Orsay (IPN Orsay, France) and used for the study of gold and platinum radioisotopes at the ISOCELE mass separator (Lee *et al* 1987b, 1988, Duong *et al* 1989). Lately, this method has been exploited at ISOLDE by the COMPLIS apparatus (Sauvage *et al* 2000) which has produced a series of IS and HFS measurements on radioisotopes of gold, platinum, iridium tin and tellurium (see table 1 for references).

A substantial reduction of Doppler broadening in photoionization laser spectroscopy is possible by applying

a collinear geometry of interaction of the laser beam with the beam of accelerated atoms (Kudryavtsev and Letokhov 1982). An experiment on ytterbium isotopes by RIS in collinear geometry with a fast atomic beam was carried out at ISOLDE by Schulz *et al* (1991). Fast ions were neutralized in a charge exchange cell and the resonance ionization from a metastable atomic state level was performed by CVL-pumped dye lasers at a 10 kHz pulse repetition rate. The efficiency of detection was 1×10^{-5} and limited mainly by the duty cycle and low population of the metastable state. However, the optical resonances were measured with typical errors of 5 MHz. This method would profit from the availability of a pulsed ion beam synchronized with ionizing lasers. This opportunity has appeared at ISOLDE since installation of the radio-frequency quadrupole cooling and bunching system (ISCOOL), which is capable of accumulating and bunching radioactive ion beams (Fränberg *et al* 2008). Following the proposal by Billows *et al* (2008) an experiment on collinear resonance ionization spectroscopy (CRIS) has been mounted at ISOLDE. At the time of writing this paper, a first CRIS on-line run on the study of francium isotopes has been carried out.

Extremely high spectral resolution is achievable with Doppler-free methods of laser spectroscopy. The two-photon anti-collinear excitation method was combined with laser resonance ionization in order to precisely measure nuclear charge radii of Li isotopes (Nörtershäuser *et al* 2003). To provide highest resolution and accuracy, the single-mode cw Ti:Sa and dye lasers were applied for the ionization of atoms evaporated from a carbon foil by a CO₂ laser. Two-photon excitation of the 2S–3S transition with the use of a 735 nm Ti:Sa laser was followed by spontaneous decay 3S–2P and by excitation 2P–3D with a 610 nm dye laser. Transitions to the ionization continuum from the 3D state could be performed by either 735 or 610 nm photons. Tuning of a Ti:Sa laser across the two-photon resonances with a frequency precision of a few tens of kHz was achieved using a frequency-offset locking technique. The measurements were carried out at the ISOL facilities at GSI-UNILAC (Darmstadt, Germany) for ^{8,9}Li (Ewald *et al* 2004) and at TRIUMF for ^{9,11}Li (Sánchez *et al* 2006).

3.2. In-source laser spectroscopy

3.2.1. Offline in-source spectroscopy. Laser spectroscopy of atomic transitions is often performed in experiments on RIMS. Measurements of the ion yield on a selected mass versus the wavelength of laser used to produce ions at the source are called ‘in-source’ laser spectroscopy. Due to a high sensitivity, this method was used extensively for the spectroscopy of rare isotopes and in particular for elements without stable isotopes which are available only in small samples.

The study of francium Rydberg levels, and the determination of its ionization threshold (Andreev *et al* 1986b, 1987), is one of the first applications of this technique.

A series of experiments on the determination of atomic ionization potentials of actinides has been performed at Mainz University using CVL-pumped dye lasers and a time-of-flight mass spectrometer for RIMS (Riegel *et al* 1993, Erdmann *et al* 1998). With the replacement of CVL-pumped dye lasers

by Nd:YAG-pumped Ti:Sa lasers, the work on spectroscopy of actinides has been continued (Grüning *et al* 2004, Raeder *et al* 2011a, b, Gottwald 2011). The same apparatus has also been used by Mattolat *et al* (2010) and by Rossnagel *et al* (2012) for the determination of the first ionization potentials of technetium and actinium via measurements of Rydberg series.

For the ultra-sensitive investigation of trans-uranic nuclides, Backe *et al* (1997) have developed a method of ion guide-detected resonance ionization spectroscopy. It combines ionization in a gas cell, an electrode system for guiding ions to a nozzle, an ion-guide gas jet and a quadrupole mass spectrometer. This compact apparatus has been applied for a first observation of atomic levels in fermium (Sewtz *et al* 2003, Backe *et al* 2005).

3.2.2. In-hot-cavity in-source spectroscopy. A new, highly sensitive method of laser atomic spectroscopy of radioisotopes produced at ISOL facilities with RILIS has been suggested and demonstrated by Alkhazov *et al* (1992). In this method, mass separated short-lived nuclides are detected by nuclear spectroscopy techniques while scanning a RILIS laser wavelength across atomic resonance. Using this technique, measurements of IS and HFS have been carried out at the IRIS/PNPI facility for neutron-deficient isotopes of ytterbium, thulium, europium, gadolinium and thallium (Barzakh *et al* 2000, 2004, 2005, 2012).

The observable widths of atomic resonances are defined by Doppler and/or pressure broadening, depending on the conditions and type of the source. However, often isotopic and HFS effects are detectable despite the line broadening if a narrow-line laser is used in a RILIS setup. The high temperature conditions of a hot cavity RILIS are unfavourable for isotopes of light elements, while for heavier ones the Doppler effect is smaller. Fedosseev *et al* (2003) demonstrated a precision of better than 100 MHz in HFS measurements of stable thallium isotopes by in-source spectroscopy at ISOLDE RILIS. In most cases, this accuracy is sufficient for the study of mean square charge radii variations along the isotopic chains.

At ISOLDE the in-source spectroscopy of HFS and IS has been applied to radioactive isotopes of silver (Marsh 2007), copper (Köster *et al* 2000b, 2011, Weissman *et al* 2002a, Stone *et al* 2008, Flanagan *et al* 2009), lead (Andreyev *et al* 2002, Seliverstov *et al* 2006, 2009, De Witte *et al* 2007), bismuth (Marsh 2007) and polonium (Cocolios *et al* 2011). In 2011, a run on in-source spectroscopy of neutron-deficient thallium isotopes was carried out at ISOLDE; the data are being analysed.

The in-source laser spectroscopy was required also for establishing the ionization scheme of polonium and astatine, which do not have stable isotopes. Using quite limited literature data, it was possible to find appropriate resonance transitions between excited states in order to find an efficient ionization scheme for polonium (Cocolios *et al* 2008). Recently, a series of experiments on in-source spectroscopy of astatine was carried out at ISOLDE/CERN and ISAC/TRIUMF. This resulted in the development of an efficient three-step ionization scheme and first-ever measurement of the atomic ionization potential of astatine (Rothe *et al* 2012).

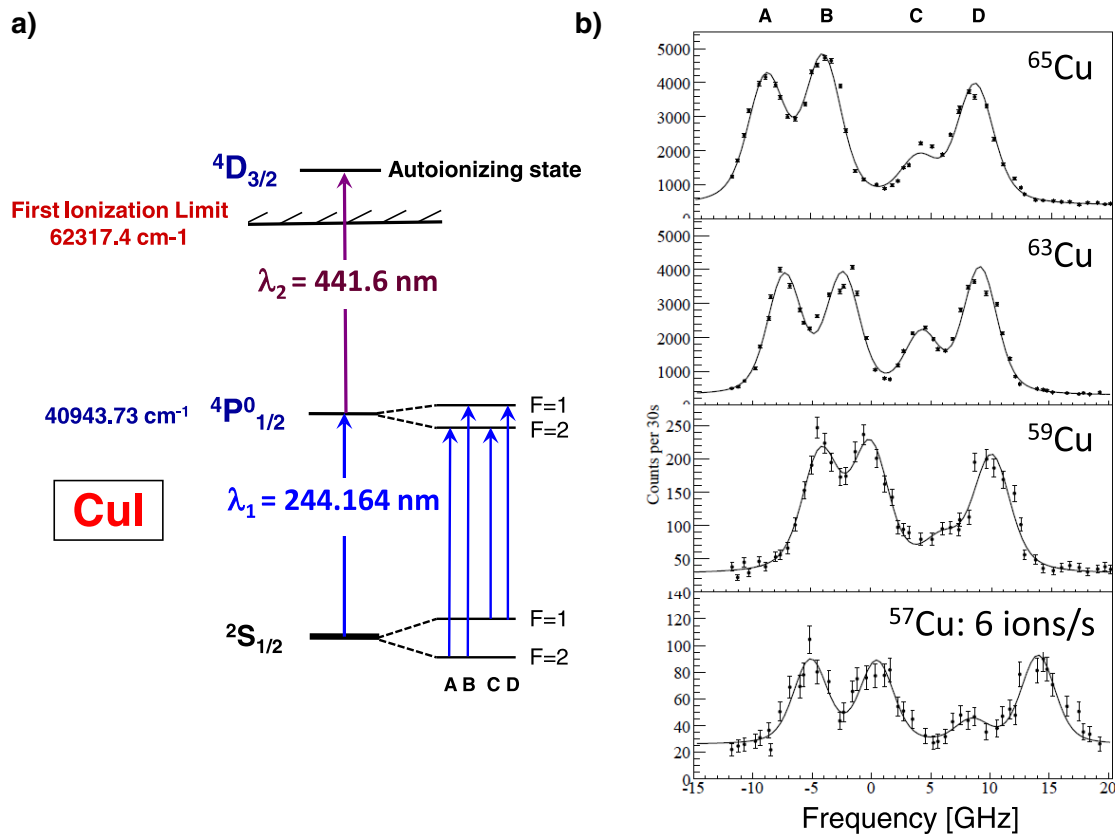


Figure 6. (a) Laser ionization scheme of copper with hyperfine splitting and (b) examples of the single spectra of $^{57,59,63,65}\text{Cu}$. Each point is sampled for 30 s. ^{57}Cu and ^{63}Cu are measured simultaneously and so are ^{59}Cu and ^{65}Cu . The frequency axis is centred at the centre of gravity of ^{63}Cu . A, B, C and D are labels for each hyperfine transition as shown in (a).

3.2.3. In-gas-cell in-source spectroscopy. The gas-cell-based laser ion source can provide high sensitivity and selectivity to study exotic neutron-deficient and neutron-rich nuclei and can be used for laser spectroscopy studies (Sonoda *et al* 2009). The application of the in-source laser spectroscopy technique has been performed at the LISOL online mass separator (Cocolios *et al* 2009, 2010a). The HFS of the odd-A isotopes $^{57,59,63,65}\text{Cu}$ as well as that of ^{58}Cu were measured. The magnetic dipole moments and changes in the mean-square charge radius have been extracted and the spin assignment for those isotopes has been confirmed. The laser ionization scheme of copper with hyperfine splitting and examples of the single spectra of $^{57,59,63,65}\text{Cu}$ are shown in figure 6. Much higher spectral resolution can provide on-line laser resonance ionization spectroscopy in a gas jet (Kudryavtsev *et al* 2012).

Resonance ionization spectroscopy in a buffer gas cell with radioactive decay detection (RADRIS) has been developed by Lauth *et al* (1992) and successfully applied for hyperfine spectroscopy of short-lived $^{240\text{f},242\text{f},244\text{f}}\text{Am}$ fission isomers (Backe *et al* 1998, 2000). A modification of this method that allows efficient collection of neutralized ions on a filament with subsequent fast evaporation and selective laser ionization has been demonstrated by Backe *et al* (2007).

3.3. In-source separation of nuclear isomers

Isomeric nuclei in general have different spins, magnetic dipole and electric quadrupole moments. These differences

produce different hyperfine splitting of atomic lines. Therefore, operating the laser ion sources using narrow-band lasers for in-source spectroscopy experiments it is possible to find conditions for preferential ionization of certain nuclear isomers.

Very soon after the first experiments on resonance laser ionization, Letokhov (1973) considered a possibility of separating nuclear isomers by laser radiation. The feasibility of his idea of ‘a laser spectrograph of the isomeric states of nuclei’ was explored for ^{197}Hg (Dyer *et al* 1985), ^{141}Sm and ^{164}Tm (Mishin *et al* 1987b). However, separated ion beams of nuclear isomers were produced for the first time at ISOLDE with RILIS during the study of silver isotopes (Kratz *et al* 1998, Fedoseyev *et al* 2000). Isomer separation was also demonstrated for $^{68,70}\text{Cu}$ (Köster *et al* 2000a, b), ^{185}Pb (Andreyev *et al* 2002), $^{183,185,187,189}\text{Pb}$ (Seliverstov *et al* 2006, 2009) and ^{195}Po (Cocolios *et al* 2010). At the IRIS/PNPI facility the isomer selectivity with RILIS has been demonstrated for ^{154}Tm (Barzakh *et al* 2000), ^{145}Gd (Barzakh *et al* 2005) and $^{185,186}\text{Tl}$ (Barzakh *et al* 2012). A separation of ^{105}Ag isomers using Ti:Sa lasers was demonstrated also at TRIUMF (Lassen *et al* 2009 and Geppert 2008).

The isomer-selective ionization substantially facilitates understanding of the nuclear structure manifested in α , β and γ decay processes. In particular, two α -decaying isomeric states have been identified in ^{185}Pb by Andreyev *et al* (2002). The isomer selectivity of the produced ion beams combined with mass measurements and β decay studies has enabled

the unambiguous identification of three β -decaying isomers in ^{70}Cu (Van Roosbroeck *et al* 2004). The first use of post-accelerated radioactive isomeric beams of $^{68,70}\text{Cu}$ for Coulomb excitation experiments was reported by Stefanescu *et al* (2007). Sauvage *et al* (2009) constructed a level scheme of ^{189}Tl based on isomer-selective ionization of ^{189}Pb . Similarly, Cocolios *et al* (2010) reconstructed the structure of ^{191}Pb from the α decay of ^{195}Po isomers.

4. Conclusion

Resonance laser ionization has proven to be a very suitable method for the production and research of radioactive isotopes. Today a laser ion source is a must for any ISOL facility. It represents the most universal means for the selective production of radioactive ion beams. Depending on the target conditions different types of sources and lasers can be used to produce ion beams efficiently. Table 1 summarizes the data on radioactive isotopes that became available for research as ions thanks to resonance ionization with lasers.

In a private conversation with one of us (VF) during the first European Conference on Atomic Physics at Accelerators (APAC 99), Vladilen Letokhov said that the resonance laser ionization method has taken a prominent position in nuclear research and deserves a review article devoted to it. We believe that through this paper we could fulfil at least partially that advice.

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